Agglomerated Fillers and Universal Composites

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Introduction

The Two Composite Problem

Before the introduction of universal composites, clinicians needed separate composites for anterior and posterior restorations. The microfillers (> 1 μm in diameter) used in posterior composites resulted in strong restorations with low volumetric shrinkage. However, as the restorations wore down through use, the larger filler particles were exposed and caused surface roughness unsuitable anterior applications. To make a more polishable and aesthetic composite, manufacturers developed nanofilled and hybrid composites with smaller fillers (< 1 μm in diameter) to wear to a smoother surface. Figure 1 shows the effect of filler size on surface roughness.

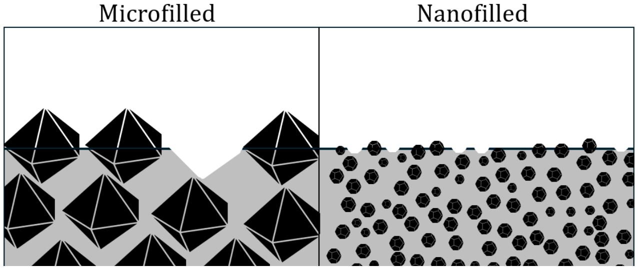

Figure 1. The difference in surface roughness of a microfilled composite vs a nanofilled composite. Fillers (black) of different sizes stick out above the surface of the resin (grey) or are removed by mechanical forces leaving holes.

Unfortunately for manufacturers and clinicians, nanofilled anterior composites had lower strength and higher volumetric shrinkage than their posterior counterparts. In a composite, the resin that coats and binds the filler particles is weaker than the filler and shrinks when cured. Smaller filler particles require more resin to bind them together, resulting in a weaker composite with increased shrinkage. To make a product with acceptable properties for both anterior and posterior use, manufacturers had to develop a filler technology that had the strength and shrinkage of a microfiller and wear like a nanofiller. Thus, the agglomerated filler was born.

Agglomerated Filler Technology

The term "agglomerated filler" or "aggregated filler" refers to any technology that loosely binds together nano-sized fillers into micro-sized bundles. Different composite manufacturers use different methods to achieve these bundles – with different features as a result. Commercial methods for manufacturing agglomerated fillers are sintering/calcination and pre-polymerization.

Sintering

Sintering involves heating nanofillers to >1000°C to fuse them together in porous clusters. These clusters have decreased surface area compared to the loose nanofillers[1,2]. The sintered fillers are then treated with an appropriate coupling agent before incorporation into a resin matrix[1]. The relatively weak cohesion formed by sintering will break when exposed to high forces, resulting in a filler that acts as nano-sized particles when exposed to wear (Figure 2). When compared to un-sintered nanoparticles, nanoclusters resulted in higher flexural modulus and fracture toughness along with lower surface roughness[2].

Pre-Polymerization

Pre-polymerization uses resin to agglomerate nanofillers before incorporation into the final product. A multi-step process is used, where nanofillers are treated with a coupling agent, mixed with resin, cured, and ground into micron-sized particles for use as microfillers in a composite[3]. When compared to sintered nanoclusters, pre-polymerized fillers (PPF) offer more flexibility in terms of composition and refractive index while retaining polishability. Materials that cannot be sintered, either because of chemical composition or because of heat sensitivity, can be incorporated into a PPF.

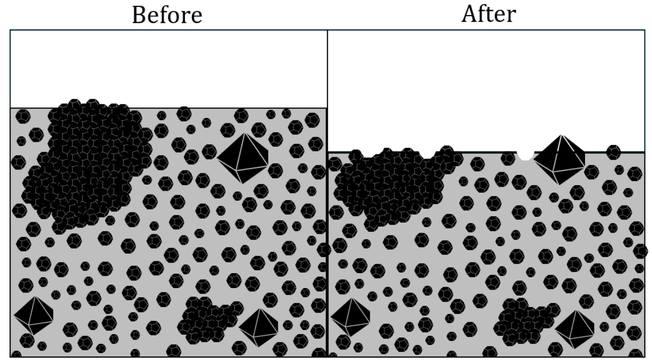

Figure 2. Comparison of surface roughness of composite containing aggregated filler before (left) and after (right) wear.

Universal Composites

Balancing Properties

For modern universal composites, an agglomerated filler is the key to optimizing the physical and mechanical properties of the final product. As discussed previously, surface roughness and volumetric shrinkage consistently work in opposition to one another. However, these are not the only two properties that traditional filler technology has linked together. Figure 3 describes some of the opposing properties manufacturers encounter when developing a universal composite.

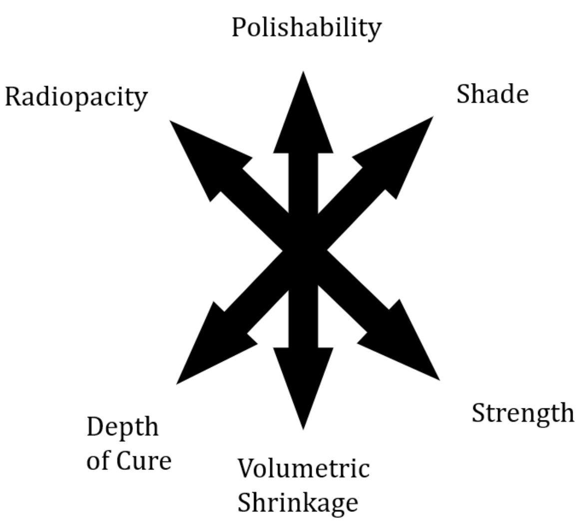

Figure 3. Opposing properties in current composite formulations.

To maximize polishability, the smallest fillers are chosen. The surface-area-to-volume ratio of these nanoparticles is very large, requiring large amounts of resin to coat the surfaces and bind them together. When the composite is cured, the resin shrinks, causing relatively high volumetric shrinkage.

Shade and depth of cure are opposed properties in light-cured materials. Since light penetration is required to cure the material, the darker the color, the less light penetrates and the lower the depth of cure.

Finally, radiopacity is opposed to a number of strength properties. Radiopaque fillers are not as strong as traditional glass fillers, so large quantities of these materials tend to have a negative effect on the overall strength properties of the composite.

Agglomerated fillers uncouple several of these linked properties. Formulators can now independently maximize opposing properties, resulting in a better finished product.

Clinical Relevance

All resin-based composites shrink as they cure. The amount of shrinkage varies based on the resin composition: resins with more double bonds per unit volume shrink more than resins with fewer double bonds[4]. This means that a resin made entirely out of BisGMA will shrink approximately 9.2% and a resin made of triethylene glycol dimethacrylate (a smaller monomer) will shrink approximately 15.5%[4]. Pre-polymerized fillers help to reduce the amount of shrinkage by pre-curing and "pre-shrinking" the resin around nanofillers, so that the shrinkage stress is not transferred to the tooth during a restoration. A study of six bulk-fill composites showed a close relationship between linear shrinkage percent and shrinkage force[5], with a 1% increase in linear shrinkage corresponding to approximately 8 N of extra force. Reducing volumetric shrinkage and the resulting shrinkage force leads to fewer gaps between the tooth and the restoration and better clinical outcomes[6,7].

The ability of agglomerated fillers to strengthen polishable composites also affects clinical outcomes. Increased flexural strength and flexural modulus can prevent the restoration from fracturing. Strength can be further increased by using a strong PPF resin, which will allow for the inclusion of additional radiopacity agent.

Higher radiopacities result in whiter spots on radiographs, making it easier for clinicians to identify previous restorations. Studies show increased levels of radiopaque filler can aid in the detection of secondary caries when the overall radiopacity is higher than that of enamel[8,9] but less than that of amalgam. Since the radiopacity of enamel is approximately 2 mm Al eq.[10,11,12], the use of agglomerated fillers can result in positive clinical outcomes.

Commercial Products

All major brands offer a commercial product that takes advantage of agglomerated filler technology. Although different agglomeration methods are used, all the universal composites in Table 1 claim superior polishability and high strength.

Table 1. Universal composites using agglomerated filler technology.

Manufacturer	Composite	Agglomerated Filler Type
BISCO	Quantium™	PP
Dentsply Sirona	TPH Spectra® ST	PP
Ivoclar	Tetric EvoCeram®	PP
Kerr	Harmonize™	S
Solventum	Filtek™ Supreme Ultra Universal	S

PP = prepolymerized filler, S = sintered filler

Agglomerated fillers perform better than nanofillers when factors such as filler loading and resin type are equal[3]. However, different products optimize for different properties, so not all universal composites perform the same. Figure 4 and Figure 5 show the volumetric shrinkage and radiopacity respectively of the composites listed in Table 1. While the values obtained from universal composites are better than could be achieved from nanofilled composites alone, the differences between universal composites are still large enough to have a clinically relevant impact.

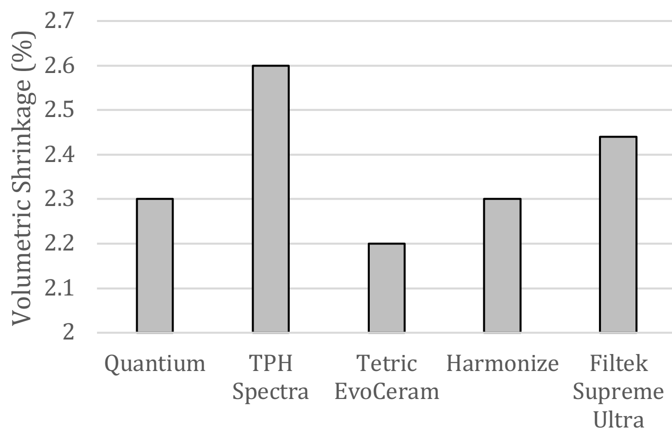

Figure 4. Volumetric shrinkage of universal composites containing agglomerated filler technology. BISCO data on file.

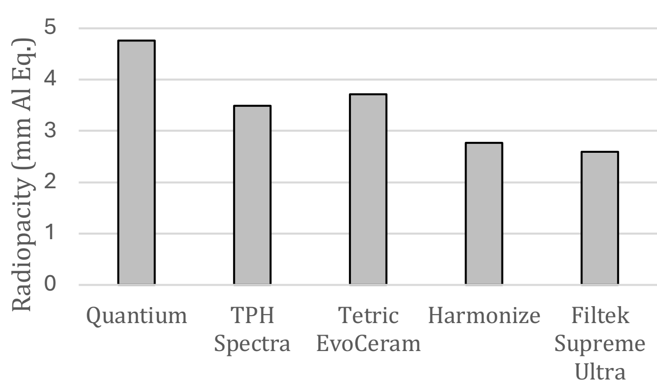

Figure 5. Radiopacity of universal composites containing agglomerated filler technology. BISCO data on file.

Future of PPF

Current Research

Pre-polymerized filler has proven its place in composites. Ongoing research continues to find new benefits as more research is published. New evidence suggests that addition of pre-polymerized filler increases fracture toughness by dissipating energy and delaying crack growth.[13]

Although their use is currently limited to composites, PPF technology is likely to expand to other restorative materials where shrinkage is problematic. Until a new resin polymerization system is widely adopted, PPFs are a good compromise between widespread compatibility and inherent propensity to shrink.

Additionally, since the PPF resin can be chosen separately from the bulk resin, differences in refractive index can be tuned to optimize translucency properties and resin strength. Traditionally, it is very difficult to increase depth of cure; however, a PPF that matches the refractive index of the bulk resin can increase translucency and therefore increase light penetration.

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